Life cycle assessment of sewage sludge treatment and its use on land

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PhD Thesis
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Preface

The work presented in this PhD thesis was conducted at the Department of Environmental Engineering of the Technical University of Denmark (DTU) under the supervision of Associate Professor Charlotte Scheutz and the co-supervision of Professor Thomas Højlund Christensen. The work was conducted from January 2011 to August 2014 and was generously funded by the 3R PhD school.

The PhD thesis is organised in five sections: an introduction presenting the background to the PhD work, three chapters summarising the findings of eight scientific journal articles, and a final section stating the conclusion and future research opportunities. The papers are referred to by their roman numerals throughout the thesis (e.g. Paper I).

I. Yoshida, H., Christensen, T.H., Scheutz, C, 2013. Life cycle Assessment of Sewage Sludge Management – a review. *Waste Management and Research* 31(11), 1083-1101.

II. Yoshida, H., Christensen, T.H., Guildal, T., Scheutz, C.. A comprehensive substance flow analysis of a municipal wastewater treatment plant. *Chemosphere*, doi:10.1016/j.chemosphere.2013.09.045

III. Yoshida, H., Mønster, J., Scheutz, C., 2014 A plant integrated measurement of methane and nitrous gas from a municipal wastewater treatment plant. *Water Research* 61, 108-118.

IV. Yoshida, H., Clavreul, J., Scheutz, C., Christensen T.H., 2014. Influence of data collection schemes on the Life cycle Assessment of a municipal wastewater treatment plant. *Water Research* 56, 292-303.

V. Yoshida, H., Nielsen, M.P., Scheutz, C., Jensen, L.S., Bruun, S., Christensen, T.H., *submitted*. Long-term nitrogen emission factors for land application of treated organic waste as organic fertiliser. *Submitted to Environmental Modeling and Assessment.*

VI. Nielsen, M.P., Yoshida, H., Scheutz, C., Jensen, L.P., Christensen, T.H., Nielsen, S., Bruun, S., Effect of sludge stabilization techniques on carbon and nitrogen dynamics and greenhouse gas emissions after incorporation of sewage sludge in soil. *Manuscript under preparation*.

VII. Nielsen, M.P., Yoshida, H., Scheutz, C., Jensen, L.S., Christensen T.H., Bruun, S. Estimation of long-term emission factors associated with land application of sewage sludge. *Manuscript under preparation.*

VIII. Yoshida, H., Christensen T.H., Scheutz, C. Life cycle assessment of sewage sludge management alternatives including long-term impacts after land application. *Manuscript under preparation.*

In this online version of the thesis, the papers are not included but can be obtained from electronic article databases e.g. via www.orbit.dtu.dk or on request from DTU Environment, Technical University of Denmark, Miljøvej, Building 113, 2800 Kgs. Lyngby, Denmark, reception@env.dtu.dk

In addition, the following publications, albeit not included in this thesis, were conducted during this PhD study period:

Yoshida, H. , K. van Dijk, A. Drizo, S.W. van Ginkel, K. Matsubae, M. Buehrer (2013). P Recovery and Reuse. Pages in K.A. Wyant, J.R. Corman, and J.J. Elser. P, Food, and Our Future. Oxford University Press, New York City, New York, USA

Yoshida, H., Clavreul, J., Scheutz, C., T.H. Christensen (2011) Development of mass flow-based Life cycle Assessment tool for sewage sludge treatment and disposal. International Water Association International Conference: Eco-technologies for Wastewater Treatment (EcoSTP), held in Santiago de Compostela, 6/25-27 2012.

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First and foremost, I would thank Dr. Thomas H. Christensen for granting me this opportunity to be part of the 3R PhD School at DTU-Environment. It was a truly unexpected twist in my life but nevertheless the best spontaneous decision I have ever made.

I express my deep appreciation to my supervisor, Dr. Charlotte Scheutz, for giving me the flexibility to explore a range of topics and at the same time keeping me on track to complete the thesis. Also, my work would not even have come close to a satisfactory conclusion without the advice and guidance of Dr. Sander Bruun, Dr. Lars S. Jensen, Dr. Anders Damgaard and Dr. Henrik Spliid.

During my PhD projects, I had the chance to work with many people in and outside of DTU. My appreciation goes to my all the co-authors: Mr. Thomas Guidal, Mr. Steen Nilsen, Dr. Julie Clavreul, Dr. Jacob Mønster and Mr. Martin P. Nielsen.

It has certainly been an intellectually stimulating time, and I owe it all to a great team of "wasters": Line, Veronica, Kos, Valentina, Stefania, Elisa, Ramona, Manfred, Davide, Roberto, Alessio, Martina, Julie, Alberto, Laura, Irina, Teme, Jacob, Marianne, Mou, Vincent, Morten and Na Yang. However, I would probably not have managed to come this far without an alternative team of "problem solvers": Anne-Harsting, Erica, Rene, Dr and Mrs Kasama and Dr Mizushima.

Despite the gloomy perceived norm of PhD life as being stressful and depressing, I truly had a surprisingly happy life at DTU and give full credit to my friends: fellow women of wastewater and the gangs of Haydi Kocum, namely Elham-R, Uli, Fabio, Carson, Ioannis, Mike, Elham-M, Gizem, Gamze, Arda, Pedram, Carlos, Anas, Pau, Borja and Katerina.

Finally, I would like to thank to my family for their unconditional love and support. Especially, I am incredibly fortunate to have such an understanding and lovable partner along with me on this journey. He gave me countless hours of reassurance and encouragement, and we went through times of joy and sorrow. Mr. Josh Gable, I devote this thesis to you.

Summary

Sewage sludge is generated as an end-product of wastewater treatment processes, and its management holds importance in the operation of wastewater treatment plants from both an economic and an environmental point of view. At the same time, the management of sewage sludge is becoming increasingly multi-focused, as renewable energy and nutrient recovery have been added to the list beyond sanitation and stabilisation of sewage sludge.

In order to organise and quantify environmental benefits and associated burdens, in order to facilitate an informed decision making process, life cycle assessments (LCAs) have been applied in the field of sewage sludge management for the past two decades. While providing a flexible platform for comparing a range of sewage sludge management options, a knowledge gap has been identified through the review of existing studies, including inconsistencies in pollutant coverage and quantification, the omission of unmetered gaseous emissions and a lack of long-term emission data regarding the land application of sewage sludge.

An LCA depends heavily on existing emission and operational data, as generating such data could be prohibitively time- and resource-consuming. Emission and operational data are already collected by wastewater treatment plants for compliance with pollutant discharge requirements, but a part of this pollutant discharge is also reported to a web-based registry (European Pollutant Release and Transfer Registry (E-PRTR)) and is available to the public free of charge. While this data source provides a standardised data collection format, its viability has been questioned due to its limited pollutant coverage and the thresholds regarding reporting requirements.

To address this issue, a targeted input data collection campaign was conducted at a municipal wastewater treatment plant. The substance flow analysis of a municipal wastewater treatment plant was conducted to identify the fate of 32 elements, and a reduction in toxicity potential was evaluated by applying USETox. The result was largely confirmative of previous studies, in that wastewater treatment is effective at removing pollutants from wastewater and concentrates them in sewage sludge.

Efforts to collect site specific emission data were also expanded to gaseous emission measurements. The tracer dilution method was applied to measure a plant-integrated emission of N_2O and CH_4 from the wastewater treatment plant. Large variations in emissions were found within and between meas-

urement campaigns, and almost ten times more emissions were found during periods of operational difficulty such as foaming or the malfunction of in-line control systems.

The LCA was based on three input data collection schemes: a compulsory environmental information disclosure requirement, a pollutant discharge monitoring requirement and state-of-the-art on-site data collection. While adequately capturing impacts in relation to global warming and marine eutrophication, an LCA based on existing data sources might underestimate impacts associated with wastewater and sludge treatment processes.

Finally, the effort to collect emission data was expanded to the use of sludge on agricultural land. The long-term consequences of sewage sludge application on land were evaluated by applying the DAISY dynamic agro-ecosystem model. The C and N mineralisation rates obtained from the 190-day laboratory-scale incubation test for sewage sludge were used to calibrate the DAISY model, and the fates of C and N in the agricultural field were simulated over a 100-year period. The outcome of the simulation was deduced further to emission factors per unit application of N fertiliser on land by fitting a linear mixed-effect model to the outcome of simulations with varying N application levels.

It was evident that the effects of inorganic N fertiliser appear immediately after its application, while improvements in crop yield and emissions of reactive N from organic fertilisers persist over time. The window of emission is dependent on the degree of stabilisation: while the effect from treated sewage sludge ceases after 25 years, the effect from the application of more stable material such as composted municipal solid waste persists over 100 years. Large variations in emission factors, due to local conditions, were observed, especially in the case of the liquid application of sludge.

When these emission factors were applied to an LCA comparing sewage sludge treatment alternatives, emissions of reactive N (NH_3, N_2O, NH_4^+, NO_3^-) into the environment were major contributors to almost all of the non-toxic impact categories. Hence, a depiction of the N balance through the target systems, beyond energy and C balance, often included in environmental assessments is vital for accurately evaluating the environmental performance of sewage sludge management options.

Dansk sammenfatning

Spildevandsslam genereres som et biprodukt af spildevandsrensning. Behandling af spildevandsslam er derfor en vigtig del af driften af rensningsanlæg set både fra et økonomisk og miljømæssigt synspunkt. Samtidig er behandlingen af spildevandsslam stadig mere aktuel, fordi vedvarende energi og genanvendelse af næringsstoffer er blevet prioriterede områder udover hygiejne og stabilisering af spildevandsslam.

I to årtier har livscyklusvurderinger (LCV) været anvendt til at kvantificere de miljømæssige påvirkninger og besparelser for behandling af spildevandsslam og med dette at kunne støtte beslutningsprocesser angående behandling af spildevandsslam. LCV giver samtidig en fleksibel platform til at sammenligne flere behandlingsmuligheder. Den manglende viden inden for feltet er blevet kortlagt ved gennemgang af eksisterende undersøgelser, herunder blev det fundet, at der er inkonsistens i forurenings-kortlægning og kvantificering, der udelades ikke-målte gasemissioner, og der mangler langsigtede emissionsdata for tilførsel af spildevandsslam på landbrugsjord.

LCV afhænger i høj grad af data for emissioner og drift. Generering af sådanne data kan være meget tids- og ressourcekrævende. Data for drift og emissioner indsamles allerede af renseanlæggene for at kunne overholde udledningskravene. En del af emissionerne af forurenende stoffer er også rapporteret til det webbaserede register (europæisk register over udledning og overførsel af forureningsstoffer (E-PRTR)), og disse data er gratis til rådighed for offentligheden. Disse datakilder har data fra standardiseret dataindsamling, men hvorvidt data kan bruges, er usikkert på grund af deres begrænsede dækning af forurenende stoffer og grænseværdier for indberetningskrav.

For at løse dette problem, blev dataindsamlingskampagnen i dette projekt udført på et kommunalt rensningsanlæg. En stof-strøms-analyse på rensningsanlægget blev udført for at identificere vejen gennem anlægget for 32 elementer og reduktionen af den potentielle toksicitet blev evalueret ved anvendelse af USETox, en konsensus-model til at karakterisere human- og økotoksicitet i forbindelse med LCV. Resultatet var stort set som tidligere undersøgelser; spildevandsrensning er effektivt til at fjerne forurenende stoffer fra spildevand og koncentrerer stofferne i spildevandsslam. Hvis kun 1% af flowet repræsenteres, er resultatet at rejektvandet fra slambehandlingsprocessen fører en betragtelig del af stof tilbage til starten af rensningsprocessen.

Indsamling af stedsspecifikke emissionsdata inkluderede gasemissionsmålinger. En metode med sporstoffortynding blev anvendt til at måle anlægsintegrerede emissioner af N_2O og CH_4 fra renseanlægget. Stor variation i emissionerne blev fundet, både i hver enkelt og imellem målekampagnerne. Næsten ti gange højere emissioner blev fundet, når der var anlægsproblemer, såsom skumning eller svigt af in-line kontrolsystemer.

LCVen var baseret på tre dataindsamlingsordninger: obligatorisk miljøoplysnings-indberetning, overvågningskrav til forureningsudledning, og state-of-art anlægs-dataindsamling. Hvis en LCV bygger på nuværende datagrundlag undervurderes miljøpåvirkningerne forbundet med spildevands- og slambehandlingsprocesserne, hvorimod påvirkningerne på marint eutrofieringspotentiale vurderes fyldestgørende.

Endelig blev data indsamlet for emissioner i forbindelse med anvendelse af slam på landbrugsjord. De langsigtede konsekvenser af spredning på landbrugsjord blev evalueret ved at anvende den dynamiske agro-økosystem-model Daisy. Mineraliserings-hastighederne for kulstof og kvælstof fra 190-dages inkubationstest for kloakslam i laboratorieskala blev brugt til at kalibrere Daisy-modellen, og skæbnen for kulstof og kvælstof på landbrugsjord blev simuleret over 100 år. På baggrund af simuleringen blev emissionsfaktorer udledt for anvendelse per enhed af kvælstof-gødning på landbrugsjord ved at sammenholde den lineære mixed-effekt model med resultatet af simuleringen med varierende kvælstof applikationsniveau.

Det var tydeligt, at virkningen af uorganisk kvælstofgødning ses umiddelbart efter spredningen, mens forbedring af høstudbytte og emission af reaktiv kvælstof fra organiske gødningsstoffer fortsætter over tid. Vinduet for emissionen er afhængig af graden af stabilisering; effekten fra behandlet spildevandsslam ophører efter 25 år, hvorimod effekten af anvendelsen af inaktivt materiale såsom komposteret husholdningsaffald varer over 100 år. Stor variation i emissionsfaktorerne, på grund af lokale forhold, blev observeret, især ved anvendelse af flydende slam.

Når disse emissionsfaktorer blev anvendt til LCV af behandlingsalternativer for spildevandsslam, var de største bidragydere, til næsten alle de ikke-toksiske påvirknings-kategorier, emissionen af reaktivt kvælstof (NH_3, N_2O, NH_4^+, NO_3) til miljøet. Derfor er beskrivelse af kvælstofbalancen gennem systemerne, udover energi og kulstofbalancen, som ofte indgår i miljøvurderingen, afgørende for præcist at vurdere den miljømæssige påvirkning fra behandlingsmulighederne for spildevandsslam.

Table of contents

1 Introduction

1.1 Background and Motivation

Sewage sludge is a solid residue generated from wastewater treatment processes. Though representing less than 1% of the total mass flow through the wastewater treatment process, the treatment and disposal of sewage sludge might account for up to 40 or 50% of the total operational costs of a wastewater treatment plant (UNHABITAT, 2008). The generation of sewage sludge has been increasing steadily, thereby reflecting the growth of a population served by a centralised wastewater treatment service and the enforcement of stricter environmental regulations (Fytili and Zabaniotou, 2008).

Sewage sludge is generally characterised by low solid content and high susceptibility to petrification.. Due to its origin, sewage sludge also contains elevated levels of pathogens. Thus, prior to disposal or land application, the sludge needs be stabilised, in order to prevent putrefaction and inactivate pathogens, and dewatered, in order to reduce sludge volume and improve transportability (Luduvice, 2007). There is a wide range of treatment processes available, and some provide the added benefit of renewable energy production. The aspect of energy recovery is particularly important for Denmark, which is aiming at completely ceasing the use of fossil carbon-base fuels by 2050 and trying to maximise the use of domestic biomass resources (DEA, 2012).

Another focal topic around sewage sludge management is nutrient recovery. Sewage sludge contains nutrients vital to plant growth and could be applied to agricultural land in place of conventional fertilisers. Europe imports phosphate ore, mainly from Russia, Morocco, Tunisia and Syria, and the recent political instability in these regions could jeopardise a stable supply of P resources in the future (HCSS, 2012). Recycling of P resources from waste and wastewater streams could be a sensible option for ensuring food security in Denmark and beyond (Cordell et al. 2009; Elser and Benett 2011).

Hence, the management of sewage sludge is often multi-focused and requires considering both economic and environmental consequences. In order to organise and quantify the environmental benefits and burden for competing management options, lifecycle assessments (LCAs) have been applied to the field of sewage sludge management and treatment in the past two decades. To ease this process, efforts have been made to develop a decision support tool specifically for wastewater and sewage sludge handling practices (Brown et al. 2010; Stokes & Horvath 2010; Pasqualino et al. 2009).

The EASETECH model is a substance flow-based LCA tool developed by the Technical University of Denmark as an extension to the LCA supporting tool for solid waste management, EASEWASTE (Clavreul et al. 2014). EASETECH traces the flow of energy and substances through target systems, in order to simulate environmental emissions.

The model comes with an extensive database of solid waste management practices which are constantly updated. The integration of sewage sludge and wastewater treatment processes into EASETECH provides several advantages by ensuring 1) a flexible and standardised data collection format for solid waste and wastewater treatment processes, 2) the ability to keep track of substance flows through the target system and 3) tools for extensive uncertainty analysis.

The present PhD project was initiated in order to expand the applicability of EASETECH beyond municipal solid waste treatment and to include another major waste stream from the urban environment, namely sewage sludge. An LCA is an extremely data input-intensive process. The development of a database and management tool for sewage sludge treatment practices would ease the burden of conducting LCAs and expand them beyond academic research and into real decision making by wastewater treatment works.

1.2 Research objectives

The objectives of this PhD thesis are to:

- Identify the key areas of data collection for sewage sludge LCA.
- Compare various environmental data-gathering schemes available for conducting an environmental assessment of wastewater and sludge treatment processes.
- Develop long-term emission factors from the application of sewage sludge on land, in order to evaluate emission changes as a result of the stabilisation of sewage sludge.

The rest of thesis is structured in three sections followed by conclusion, as follows:

- Section 2 identifies data gaps in current environmental impact assessments of sewage sludge management (Paper **I**).

- Section 3 presents potential approaches to inputting data collected when conducing LCAs of wastewater and sewage sludge treatment processes (Papers **II**, **III** and **IV**).
- Section 4 describes an environmental assessment of the land application of sewage sludge, including long-term emissions (Papers **V, VI, VII** and **VIII**).
- Section 5 presents final conclusions and recommendations for future work.

2 LCA of sewage sludge management and current data gaps

This chapter presents the LCA concept and a review of existing LCA studies on sewage sludge management with the purpose of identifying current data gaps.

2.1 Life cycle Assessment

An LCA is a framework used to quantify potential environmental impacts of targeted systems and to provide a basis for informed decision making. In essence, an LCA involves the deliberate process of quantifying, organising and translating environmental emissions into one or multiple indicators of environmental impacts. LCAs have been applied extensively for the environmental assessment of solid waste management (Pires et al. 2011; Laurent et al. 2014), and the application has also been extended to the area of wastewater treatment (Corominas et al. 2013).

The LCA process is divided into four steps, as established in line with ISO standards (2006). An LCA starts with the establishment of a goal and a scope, where the purposes of the assessment, the system boundaries and the functional units are defined and documented. Next, a life cycle inventory (LCI) is developed to follow the goal and scope of the assessment. The LCI contains environmental emissions and energy and material in-/outputs from the targeted systems. The results of the LCI are classified into selected impact categories, and then they are combined with the characterisation of factors unique to emissions and pathways (a life cycle impact assessment (LCIA)). Often, the results of the LCIA are adjusted relative to reference values (normalisation). When priorities are given to certain impact categories, the normalised results are adjusted through a weighting process. Finally, the outcome of the assessment is interpreted, in order to facilitate informed decision making.

An LCA provides a flexible platform for compiling environmental information of management alternatives which cannot be compared directly. Under the framework of the LCA, system boundaries can be extended to include upstream emissions from the provision of energy and materials. The effect of avoided emissions via the substitution of energy and material production can be included by expanding system boundaries to downstream processes and subtracting the emissions associated with the provisioning of energy and material production that otherwise occurs.

Despite its potential, the LCA has some limitations. Figure 1 illustrates the categorisation of environmental impacts in an LCA context, by expanding the classification presented by Martinez-Blanco et al. (2013). Since not all the effects exerted by the target systems fulfil these criteria, the outcome of an LCA will entail systemic underestimation. Environmental impacts and benefits from the target system are accounted for in an LCA only when all criteria are met: effects exerted from the target system, such as the emission of pollutants into the environment, are proven, the effects are quantifiable and measurable, characterisation factors are available to connect the emission of pollutants to potential damage to the environment and the impacts are included in the goal and scope of the assessment.

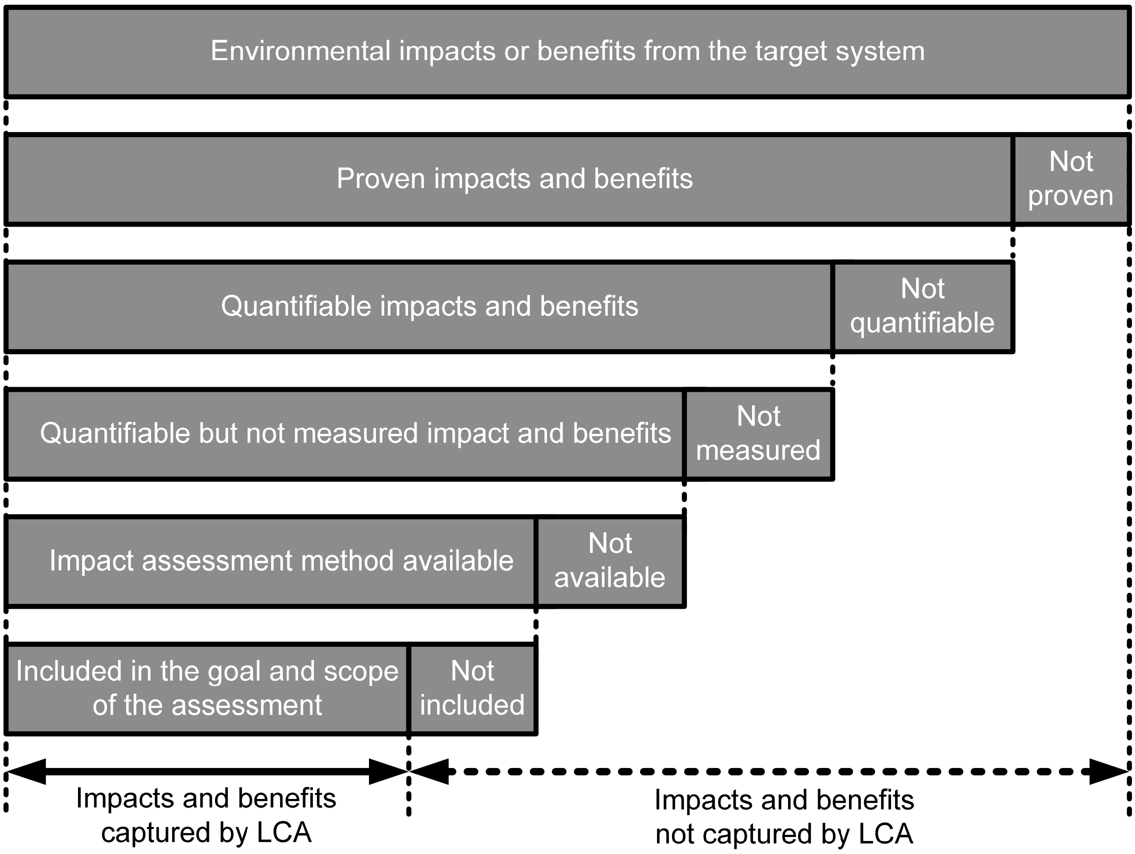

Figure 1. Criterial for impacts and benefits to be included by LCA

However, one should keep in mind that the goal of an LCA is not to capture perfectly any impacts exerted from the target system. As any kind of attempt to construct a model involves the abstraction of reality, it is impossible to capture all epistemic uncertainties that are the result of knowledge gaps; rather, the LCA provides the best attainable estimation of environmental impacts for facilitating the decision making process, by either comparing alternatives or bench-marking environmental performance. What makes an LCA viable is the constant revision and updating of the coverage of emission and impact assessment methods, based on state-of- the-art knowledge of the target systems.

2.2 LCA for sewage sludge management

A review of existing LCA studies was conducted to assess the state-of-the-art knowledge on the environmental impacts on and benefits of sewage sludge treatment and its utilisation and to identify knowledge gaps (Paper I). In total, 35 articles published in 13 international scientific journals were reviewed. The drivers and goals of the studies ranged from the selection of unit processes to the formulation of national policies. The focal topics of the assessment were the recycling of nutrients back into agricultural land, renewable energy production and trade-offs between more stringent wastewater treatment requirements and energy and treatment chemical needs.

The studies reviewed were generally conducted using a process-based approach in which the impacts were accounted for by combining plant- or process-specific operational and emission data and LCIs data taken from databases, such as Ecoinvent, BUWAL and IDEMAT for background systems. Murray et al. (2008), Hong et al. (2009) and Stokes and Horvath (2010) used a hybrid LCA to combine process-based LCAs with economic input and output LCAs. The functional unit was generally defined as the treatment of a certain volume or mass of wastewater or sludge, though some studies considered the sludge treatment process and set as the basis of comparison the production of commercially available products such as Portland cements, steam and P recycled back onto agricultural land (Hong and Li, 2011; Liu et al. 2011 and Linderholm et al. 2012, respectively).

Based on the review, it was evident that large discrepancies exist in the selection of pollutant coverage and emission pathways, and clear knowledge gaps were identified in the following areas:

Coverage of pollutants: No consensus was found regarding pollutant coverage in the reviewed studies. For instance, when evaluating the effect of heavy metal introduction to soil via the land application of stabilised sludge, the number of elements included in the studies ranged from one to 11. Cd was included all the studies but the choice of other metals was left to the LCA practitioners to decide. Similar trends were also evident when accounting for pollutant emissions into the air following on from land application (zero to 11 substances), sludge incineration (zero to 18 substances) and biogas combustion (zero to 5 substances).

Unmetered gaseous emissions: Global warming potential (GWP) was one of the most common impact categories included in the LCAs of sewage sludge management. Due to its fugitive nature of emission, on-site measurement of unmetered emissions is rarely performed and often ignored from the assessment. The leakage of methane from anaerobic digesters is one of such emissions. The inclusion of methane emission Out of 28 studies analysed anaerobic digestion process, only half included the emission of methane to the atmosphere. The inclusion of methane leakage could make the net GWP of the target system from negative (saving emissions) to positive. Yet, the rate of emission was taken from the greenhouse gas (GHG) accounting guidelines, not specific to the target systems. .

Long-term emissions from sludge application on land: Land application was included in 28 out of the 35 reviewed studies. Land application of sludge was either accounted for as the result of avoiding the production of conventional fertilisers or the introduction of pollutants to arable land. 14 studies included some sort of gaseous emission (e.g. CH_4, N_2O, NH_3), only four considered nutrient leaching and runoff. When emission of reactive N species were included, UOL was found to be the major contributor to global warming, eutrophication and acidification (Johansson et al. 2008; Peters and Rowley 2009; Brown et al. 2010; Hospido et al. 2010). Furthermore, due to lack of emission data, none of the previous studies assessed long-term consequence of land application of sewage sludge on land beyond carbon sequestration.

3 Data Source and Data Coverage

3.1 Input data sources for life cycle assessments

The quality of an LCA depends highly on the availability of input data for constructing an LCI. Completeness, representativeness and accuracy were identified as three aspects of data quality, and representativeness could be further defined in temporal, geographical and technical terms (Huijbregts et al. 2001). Nonetheless, collecting a site-specific input dataset could be prohibitively time- and resource-consuming. As a consequence, LCA practitioners rely heavily on existing emission and operational data for target systems.

At wastewater treatment plants (WWTPs), some environmental data are collected and reported in order to obtain a discharge permit, while other emissions are monitored continuously for process optimisation purposes. Furthermore, in order to ensure a public access to environmental data, the European Union established a web-based register in 2003 called the European Pollutant Release Transfer Register (E-PRTR). It is mandatory for economic entities, including WWTPs, to report their emissions every three years. The E-PRTR collects emission data from over 28,000 facilities and is designed to cover over 90% of industrial emissions of 91substances (Wursthorn et al. 2011).

The utilisation of these existing data sources could reduce the burden of input data collection and help expand the application of LCAs for decision making processes. Particularly, the adoption of compulsory disclosure requirements such as E-PRTR would allow the public access to the standardised emission data across industrial sectors and with a regular updates, which has been identified as an area of research development for LCAs (Finnveden et al. 2009). However, in both cases, there is a limitation in relation to pollutant coverage. Not all the pollutants known to cause adverse health and environmental impacts are regulated, and reporting on the subject is not mandated. Even when a pollutant is included in a list of pollutants under compulsory disclosure requirements, it is still subject to several layers of limitations, mainly by the reporting threshold set by the rule. Figure 2 illustrates the selection process for E-PRTR.

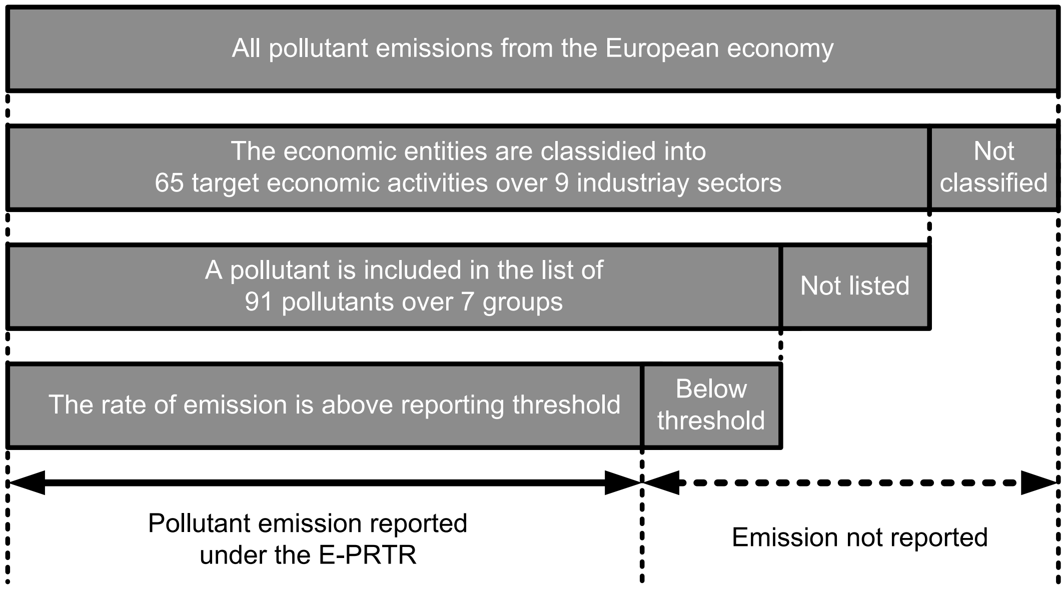

Figure 2. Selection of pollutant data for the European Pollutant Release and Transfer Registry

To evaluate the extent to which input data sources affect the outcome of an LCA, the Avedøre WWTP, located to the southwest of Copenhagen, Denmark, was used as a case study. Extensive data collection was conducted between 2011 and 2013. LCAs were conducted at three levels: based on the E-PRTR reporting guideline (L1), emission monitoring requirements under Danish regulations (L2) and emission data from a state-of-the-art measurement campaign (L3). Table 1 presents pollutant coverage in relation to three input data sources. A further description of the emission measurement campaign is presented in sections 3.2 and 3.3, and the results of the LCA are highlighted in section 3.4.

3.2 Substance flow analysis of a wastewater treatment plant

In order to provide the basis for a process-based LCA, a substance flow analysis was conducted for total mass, total solid, volatile total organic carbon, 32 elements (Al, Ag, As, Ba, Be, Br, C, Ca, Cd, Cl, Co, Cr, Cu, Fe, Hg, K, Li, Mg, Mn, Mo, N, Na, Ni, P, Pb, S, Sb, Se, Sn, Sr, Ti, V, Zn) and four groups of organic pollutants (linear alkylbenzene sulphonates, bis(2-ethylhexyl)phthalate, polychlorinated biphenyl and polycyclic aromatic hydrocarbons) (Paper II).

Figure 3 presents a schematic diagram of the Avedøre WWTP. The plant treats 25.3 million m^3 of wastewater annually, 85% of which is domestic in origin and the remainder is from industrial sources. Incoming wastewater is first treated with a bar screen and in a grit chamber, where sand is settled and fat and grease are skimmed off. The wastewater is then sent to the primary sedimentation tank and then for secondary treatment through biological nitrogen removal. To ensure the adequate removal of P, iron salts are added before the effluent reaches the secondary clarifiers. Both primary and secondary sludge is sent to anaerobic digestion. The digested sludge is then dewatered by a centrifugal machine and sent to an incinerator, while the rejected water is sent back to the head of the treatment works. Ash captured by bag filters and electrostatic precipitators is deposited in an on-site landfill.

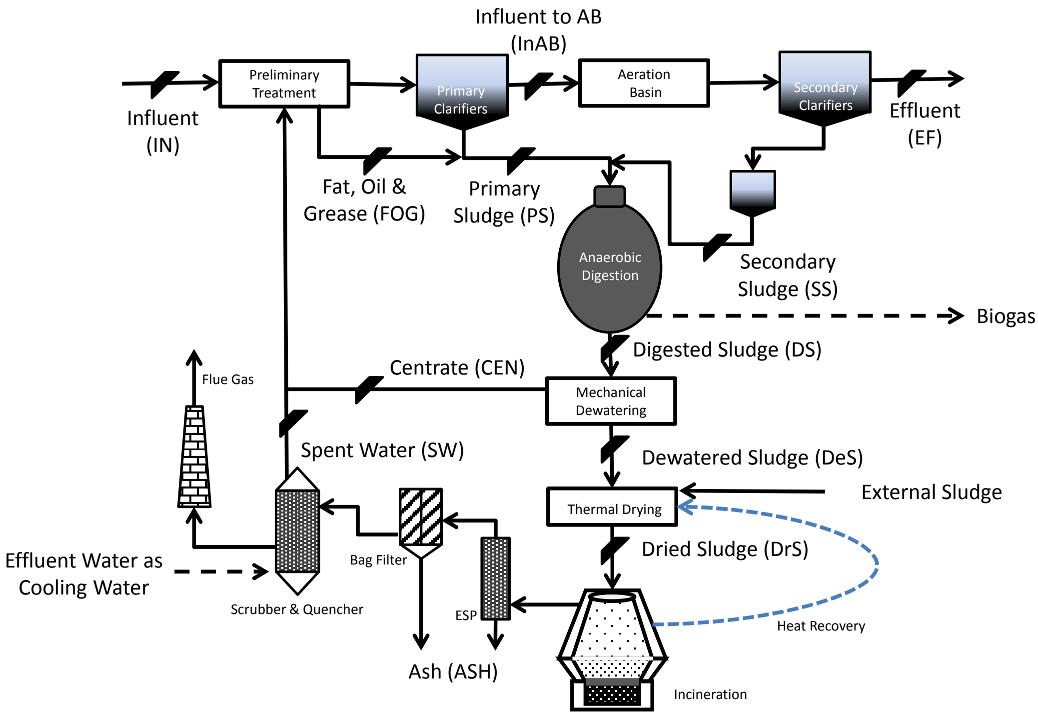

Figure 3. Schematic diagram of the Avedøre Wastewater Treatment Plant. The tilted trapezoid indicates the sampling point (Paper II).

The mass flow analysis was conducted by using the STAN (subSTance flow ANalysis) model developed by the Vienna University of Technology. STAN calculates the best fitted values iteratively, by using a successive linear data reconciliation process and Gauss's Law of error propagation (Cencic and Rechberger, 2008). In general, the mass balances presented good closure: 88% of flows characterised in the study were accompanied by an uncertainty range less than 30% of the mean values. Gaseous emissions tended to have large uncertainty ranges, though, since they are often calculated as the difference between influent and effluent flows.

The fate of substances during wastewater and sludge treatment processes is presented in Figure 4. It is evident that both inorganic and organic elements accumulated in the sewage sludge, with the exception of elements that are highly soluble or degradable by wastewater and sludge treatment processes. The majority of metals and metalloids were further accumulated in the incineration ash, while organic pollutants were effectively destroyed – both biologically and thermally.

Side streams from the sludge treatment process back to the wastewater treatment, such as rejected water from the dewatering process, carried less than 1% of volume flow back to the head of the treatment process, but they nevertheless represented sizeable substance flows. In the case of P, as much as 28% of P entering the wastewater treatment processes was introduced from water rejected from the mechanical dewatering of sewage sludge.

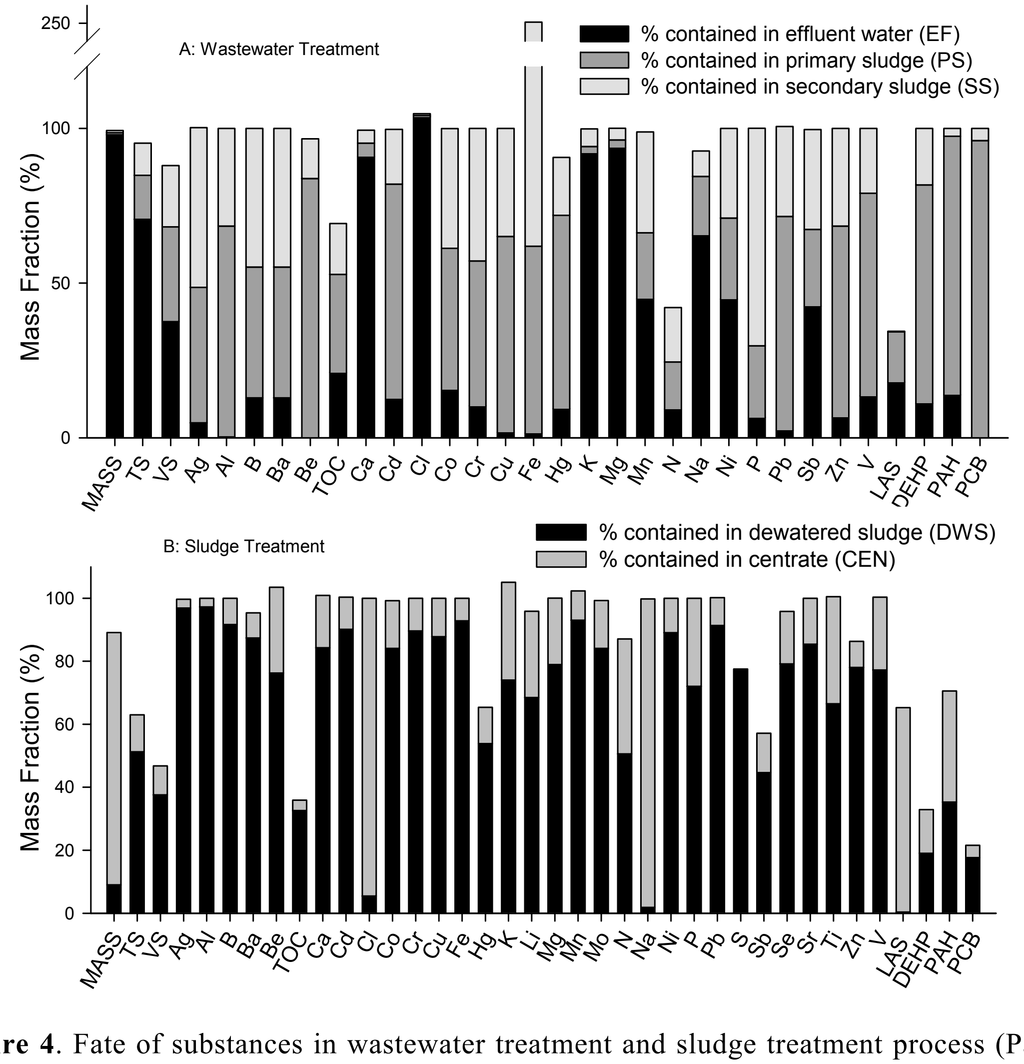

Figure 4. Fate of substances in wastewater treatment and sludge treatment process (Paper II).

The screening of human and eco-toxicity impacts following on from effluent discharge and the land application of sewage sludge was conducted by applying the USEtox consensus-based toxicity characterisation tool. The results showed that removing inorganic constituents reduced toxic impact potential associated with incoming wastewater by 87-92%.

3.3 Gaseous emissions from wastewater treatment plants

Besides the discharge of pollutants into surrounding water bodies, WWTPs contribute to anthropogenic greenhouse gas (GHG) emissions into the atmosphere. The quantification of GHG emissions from wastewater treatment plants has so far been conducted mainly by using a floating flux chamber or by utilising existing air collection systems. The former method only provides a spot measurement of GHG emissions, while the latter cannot capture structural leakages or fugitive emissions.

A tracer dispersion method was applied to quantify plant-integrated, real-time emissions of CH_4 and N_2O from the Avedøre WWTP (Paper III). Two mobile cavity ring down spectroscopy sampling devices were used to record downwind concentrations of gases emitted from the WWTP. The rate of emission was determined by comparing the air concentration of the target substance with the concentration of a tracer gas, which was emitted at a constant rate at the plant. Figure 5 presents the plumes of CH_4, N_2O and tracer gas detected.

In total, nine measuring campaigns were carried out. A wide range of emissions were detected: 4.99 – 92.3 kg CH_4 h^{-1} and under the detection limit (0.37 kg h^{-1}) and up to 10.5 kg N_2O h^{-1}. The emission of GHGs appeared to be highly localised, in that CH_4 was primarily emitted from the anaerobic digesters, whereas N_2O was primarily emitted from the secondary treatment process involving biological nitrogen removal. Correlating these emission rates to operational parameters, CH_4 emissions detected during the measurement campaigns corresponded to 2.07-32.7% of the CH_4 generated in the plant. As high as 4.27% of nitrogen entering the WWTP was emitted as N_2O.

High emissions were observed during periods experiencing operational problems. High emissions of CH_4 were observed when the plant was experiencing a foaming problem and the anaerobic digesters were vented to prevent excessive pressure build up. The elevated emissions of N_2O were detected along with the accumulation of nitrate in the biological nitrogen removal process, which indicates insufficient denitrifier activity.

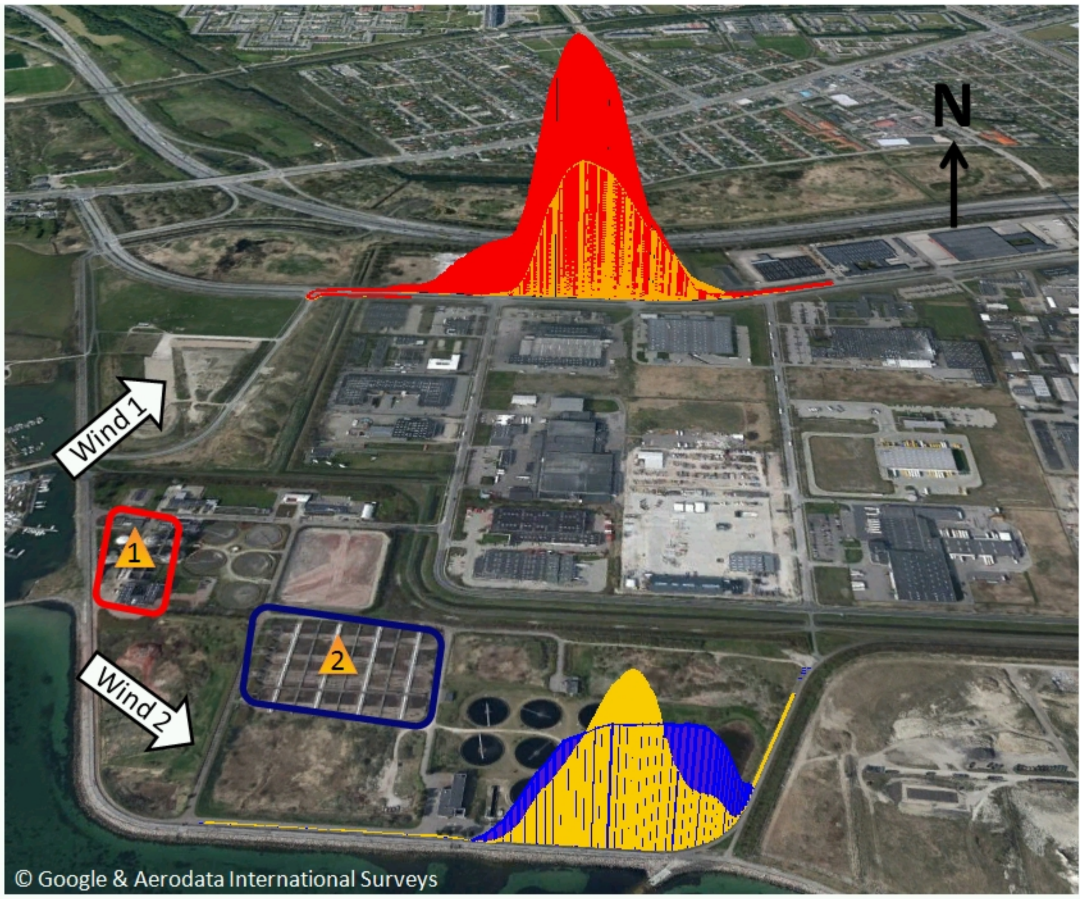

Figure 5. Methane (red), nitrous oxide (blue) and ethylene plume (yellow) detected at the wastewater treatment plant (Paper III). A red square indicates the location of anaerobic digesters and a blue square for aeration basins. Yellow triangles are where ethylene was released.

3.4 Comparison of Input Data Sources

An LCA was conducted for Avedøre WWTP to illustrate the influence of the selection of input data sources on the outcomes of an environmental impact assessment (Paper IV). The functional unit of the assessment involved the treatment of average daily inflows of wastewater at the Avedøre WWTP.

Four levels of input data collection were done in three parts: coverage of pollutants by E-PRTR (L1), monitoring requirements (L2) and state-of-the-art data collection conducted at the Avedøre WWTP (L3). In addition, emissions from background systems providing, for instance, electricity and treatment chemicals were included in L3+.

Parameter uncertainty was propagated through Monte Carlo simulation with 100,000 simulation runs. For L3+, uncertainty due to variations in electricity and treatment chemical usage on site, uncertainty due to data quality (completeness, temporal correlation, geographical correlation, technical correlation and reliability based on the work of Weidema (2012) and uncertainty embedded in the LCI dataset used to model the background system were also assessed.

Figure 5 presents the probability density profile for each data collection scheme for the nine impact categories assessed in this study. While more than 90% of non-carcinogenic human toxicity and marine eutrophication potential were captured by the current data collection scheme, due to the threshold on reporting the E-PRTR would not fully capture the impact for particulate matter emission, terrestrial acidification and terrestrial eutrophication. An LCA based on a, existing data collection scheme underestimates environmental impact potential because of limitations in substance coverage, and it is also necessary to conduct a site-specific measurement campaign, especially when looking at unmetered gaseous emissions.

Besides differences between data collection schemes, the results showed that 3-13,500% of the impacts came from background systems, such as those providing fuel, electricity and chemicals, which do not need to be disclosed currently under the E-PRTR. The incidental release of pollutants was also assessed by employing a scenario-based approach, the results for which demonstrated that these non-routine emissions, such as elevated N_2O and CH_4 during operational difficulties, could increase overall WWTP GHG emissions by between 113 and 210%.

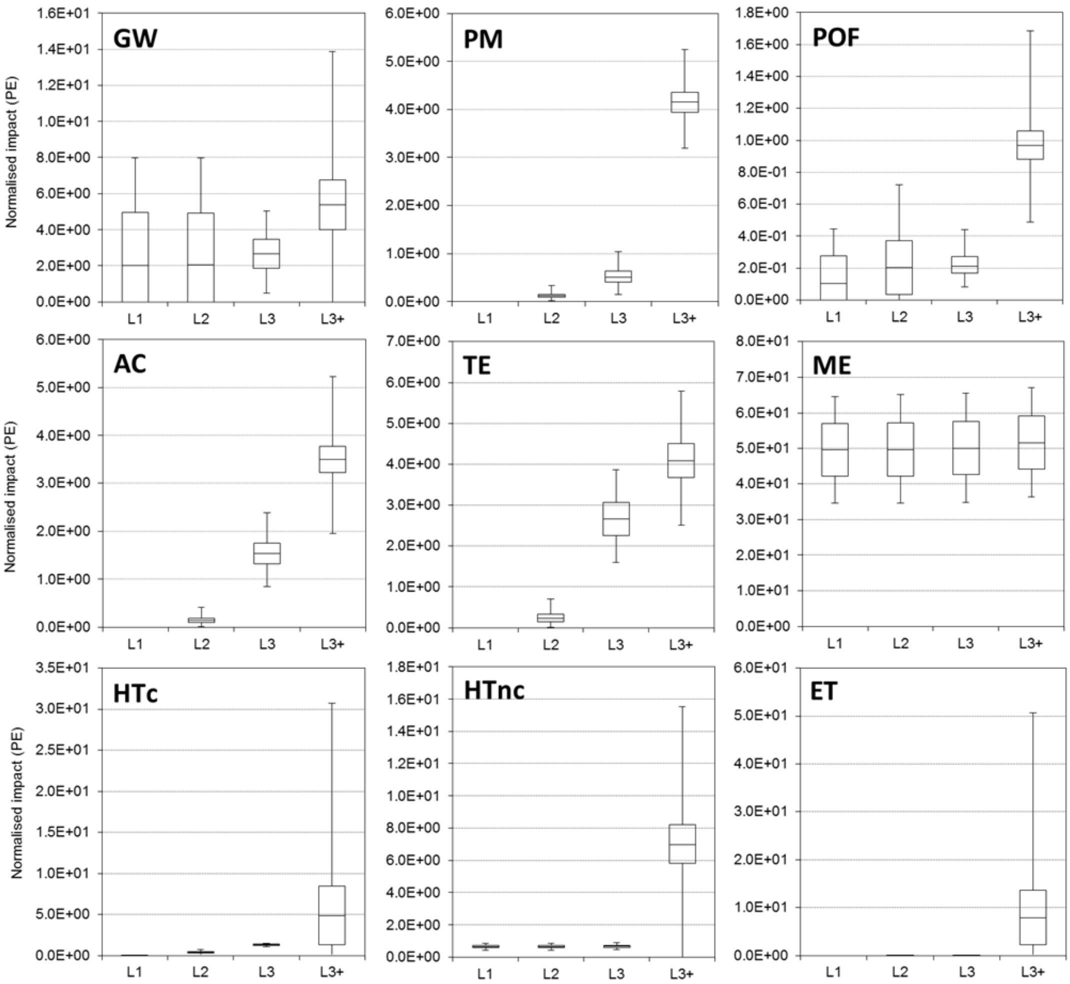

Figure 5. Normalised impacts obtained for data collection schemes L1, L2, L3 and L3+ (Paper IV).The boxes represent the 25% and 75% percentiles as well as the median, while the error bars show the minimal and maximal values obtained (GW: global warming; AC: acidification; TE: terrestrial eutrophication; ME: marine eutrophication, marine; POF: photochemical ozone formation; ET: ecotoxicity; HTc: human toxicity, cancer; HTnc: human toxicity, non-cancer; PM: particulate matter).

4 Land application of organic fertiliser

4.1 Closing the nutrient cycle

The stabilisation of organic waste yields end-products which can be applied to agricultural land as soil conditioner and fertiliser. This closes the nutrient cycle between the site of production and the site of consumption. In addition, organic fertiliser brings organically bound carbon to the soil, some parts of which can remain in the soil for prolonged periods (i.e. 100 years). This storage of carbon in the soil is largely considered to reduce CO_2 emitted into the atmosphere and mitigates global climate change (e.g. Lal 2004).

A considerable portion of N in organic fertiliser is organically bound and made available to plants over time, as it mineralises and is released into soil in inorganic form. The timing of N mineralisation and the release of inorganic N in soil play a vital role in determining the fate of N in agricultural systems. The accumulation of inorganic N in soil outside the plant growth period will create conditions that may be prone to the risk of N leaching. It is well known that the rate of N mineralisation is dependent on local conditions (soil type, precipitation pattern, temperature, etc.), as well as the stabilisation technology applied to produce organic fertiliser (i.e. Cabrera et al. 2005).

A number of dynamic agro-ecosystem models have been developed to capture the effect of local conditions and differences in fertilisers properties in relation to the fate of N (e.g. DeNitrification-DeComposition (Li et al. 1992), CERES-EGC (Gabrielle et al. 2006), DAISY (Hansen et al. 1991)), but the outcome of the simulation was affected by the number of input parameters, and it was therefore hard to establish a linear relationship between the amount of fertiliser applied and emissions into agricultural systems, which are assumed in an LCA. One way of circumventing this problem is to conduct a statistical meta-modelling on the outcome of dynamic eco-agricultural modelling based on a wide range of application scenarios. In this case, approximating the outcome of the simulation with a linear regression model allows for the development of simple emission factors, thereby correlating the total amount of N fertiliser application on land and crop yield responses as well as emissions of N into the environment.

This technique has been used before to evaluate legislation on N fertiliser application (Børgesen et al. 2001) or to integrate the environmental assessment into the European-wide agro-economic assessment model (Britz and Leip 2009). The following sections present the attempt to develop emission factors for organic fertilisers derived from waste treatment, such as stabilised municipal solid waste and sewage sludge, by applying the statistical meta-modelling approach.

4.2 Derivation of emission factors

4.2.1 Description of the DAISY model

The one-dimensional version of the DAISY model (version 5.14) was used for this study. DAISY is an open soil-plant-atmosphere model developed by Hansen et al. (1991), and it consists of several sub-models which simulate the fate of water, C and N in an agricultural system. Driving variables, which have to be supplied for the simulations, are weather data (air temperature, precipitation and global radiation), soil data (soil type and depth of the soil layer) and management data (fertiliser type and application rate, crop rotation and timing for sowing, ploughing, fertilising and harvesting).

Figure 6 illustrates the major pathways of C and N as modelled by DAISY, which simulates the mineralisation of N and C with a soil organic matter (SOM) model. Organic matter is partitioned into six pools with two added types of organic matter (slow and fast turnover), two soil microbial biomasses (slow and fast turnover) and two SOM pools (slow and fast turnover). Organic material such as organic fertilisers applied to soil is portioned mainly to the added organic matter pool, with a small part of less degradable organic material in some cases going directly into the SOM pools. Partitioning between the pools, the turnover rates and the C/N ratio of the pools reflect the degradability of N and C compounds in organic material.

Organic N is turned over in conjunction with added C and gradually mineralises into inorganic N. Some inorganic N in the soil column is taken up by crops, while the remaining inorganic nitrogen is either emitted into the air as N_2 and N_2O, via nitrification and denitrification processes, or transported out of the soil through surface runoff and leaching into the subsoil. The vertical movement of nitrate and ammonium is simulated through a complicated solute transport model, including matrix flow (using the convection-dispersion equation) and macro transport through fractures and biopores.

The SOM model parameters for different waste and fertiliser types are calibrated against a range of long and short-term mineralisation studies (e.g. Luxhøj et al. 2007; Bruun et al. 2002). C and N dynamics of composted and anaerobically digested organic fractions of municipal solid waste have been characterised by Bruun et al. 2006.

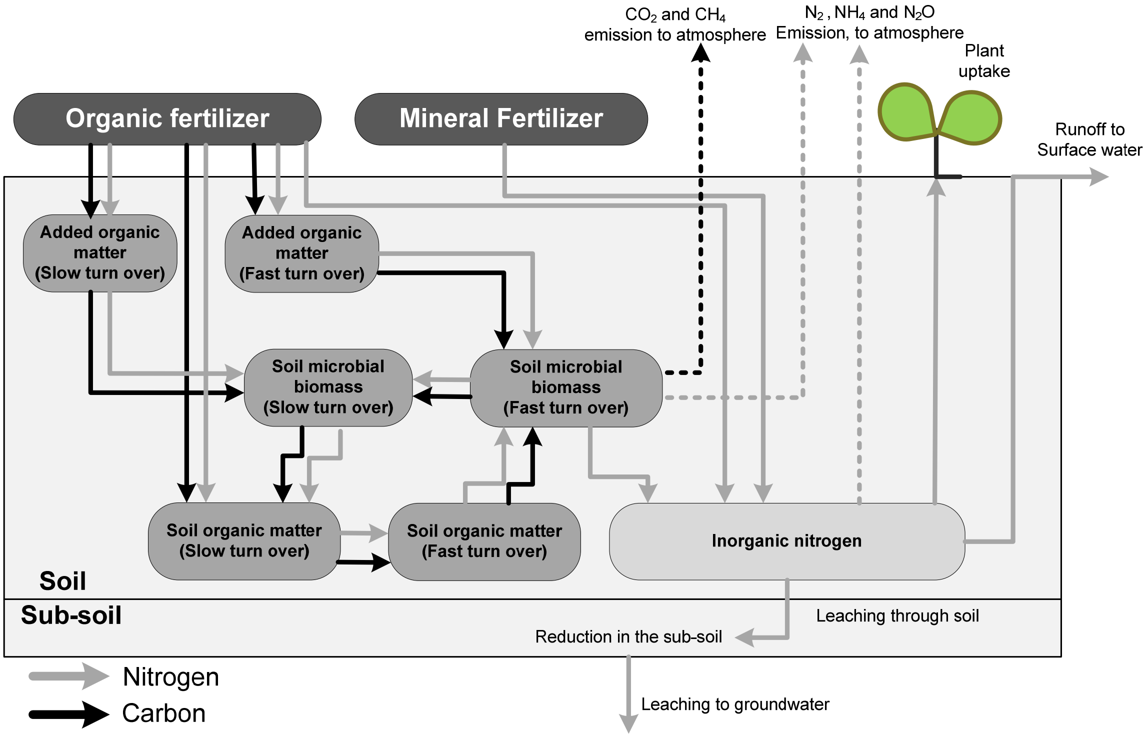

Figure 6. Schematic of the DAISY model (Paper VI)

4.2.2 Characteristics of organic fertiliser

A wide range of N turnover rates have been reported for sewage sludge (i.e. Parker and Sommers 1983, Smith and Durham 2002, Gilmour et al. 2003, Gil et al. 2011). In general, raw sludge with high and readily available organic matter content, such as primary sludge, has a lower N mineralisation rate, but degradation of readily available organic carbon through anaerobic digestion or other stabilisation measures increases the amount of organic N to be released after soil incorporation. However, more extensive means of stabilisation, such as composting, result in the reduction of N mineralisation rates via converting organic matter into a more inert form. Apart from the accumulation of knowledge on the N mineralisation rates of various sludge products, only a limited number of studies report C and N mineralisation rates simultaneously.

The simulation by DAISY of the fate of organic material requires the input of both C and N turnover rates. In order to fill this knowledge gap, a 190-day soil incubation study was performed under the incubation temperature of 15 °C, in order to better represent soil temperature in northern Europe. Twelve sludge and two mineral fertilisers were included in the incubation study (Paper IV). Table 1 tabulates C and N mineralisation rates after a 190-day incubation period. The outcome of the study largely confirms the previous studies, in that stabilisation measures reduce the C mineralisation rate but increase the N mineralisation rate, except in the case of extensive process such as composting. To the author's knowledge, this is the first time reed bed sludge has been the subject of a soil incubation study, even though approximately 10% of sludge generated in Denmark is treated in such manner. The outcome of the study shows that when treated over a full cycle (eight years), the C and N mineralisation rates of reed bed sludge fall in between anaerobically digested sludge and composted sludge.

Table 1. C and N turnover rates after 190 days of soil incubation.

Mineralisation Rate	Nitrogen (% Organic N added)	Carbon (% Organic C added)
Primary Sludge	2%	91%
Dewatered Secondary Sludge	39%	59%
Dewatered Mixed Sludge	34%	54%
Limed Sludge	46%	39%
Anaerobically Digested Sludge	7%	53%
Dewatered Anaerobically Digested Sludge*	33%	53%
Dewatered Anaerobically Digested Sludge*	47%	36%
Dewatered Anaerobically Digested Sludge*	50%	36%
Dried Anaerobically Digested Sludge	40%	57%
Reed Bed Sludge Accumulated over 2 years	40%	84%
Reed Bed Sludge Accumulated over 8 years	25%	15%
Composted Anaerobically Digested Sludge	10%	4%

*Samples were taken from three different wastewater treatment plants

4.2.3 DAISY simulation setup

In order to calculate environmental emissions resulting from the application of waste-derived fertilisers on agricultural land, the results of the soil incubation study are extrapolated for 100 years, with a warm-up period of 16 years. In all simulations, an eight-year crop rotation (spring barley, winter wheat, winter wheat, winter barley, winter rape, winter wheat, winter wheat, maize) scheme was adopted, based on an average area for each crop grown in Denmark. Maize was included, in order to reflect the recent increased production seen in northern Europe (StatBank Denmark, 2012). Standard parameterisations of crop and management dates for fertiliser application, tillage, sowing and harvesting were used (Styczen et al. 2004).

Three soil types (coarse sandy soil, sandy loam soil and heavy clay soil) and precipitation regimes (average Danish, German and Dutch precipitation regimes) were considered. For each of the fertiliser types, 12 levels of available inorganic N fertilisation rates (0, 30, 60, 90, 120, 150, 180, 210, 240, 270, 300, 330 kgN-min ha^{-1}) were simulated for both inorganic and organic fertilisers. In order to capture yearly random variability in weather data, eight sets of weather data were created by changing the starting year from 1960 to 1967 and then permutated with an eight-year crop rotation regime.

In total, the combinations of fixed and random variables resulted in 6,912 simulations (3 soil types × 3 precipitation regimes crop × 12 fertilisation levels × 64 random climate/crop combinations in the application year) for each fertiliser type, which in turn were used to make a meta-dataset. Throughout the 116 years of the simulation period, mineral fertiliser was applied according to the maximum level specified by the Danish fertiliser regulations for each crop, except for the year following the 16-year warm-up period. On the 17th year, the fertiliser application rate was modified to assess the effect of N fertiliser level on crop yields and the emission of reactive N into the environment. The simulation was kept on for another 99 years in line with the level of inorganic N fertiliser application recommended by Danish regulations.

4.2.4 Simulation outcomes

Figure 7 presents the response surfaces of average crop yields, nitrate leaching and nitrous oxide emissions for 12 fertiliser application levels over a 100-year simulation period. Responses to mineral fertiliser appear immediately after application, while the effect increases gradually over time for MSW compost, in which approximately 90% of N is present in an organically bound form. MSW digestate were assumed to be liquid applied, which means that 50% of N present in immediately plant available NH_4^+ form. As a result, the MSW digestate exhibited the characteristics of both inorganic and organic fertiliser: a quick rise at the beginning followed by a gradual increase in harvests and emissions over time.

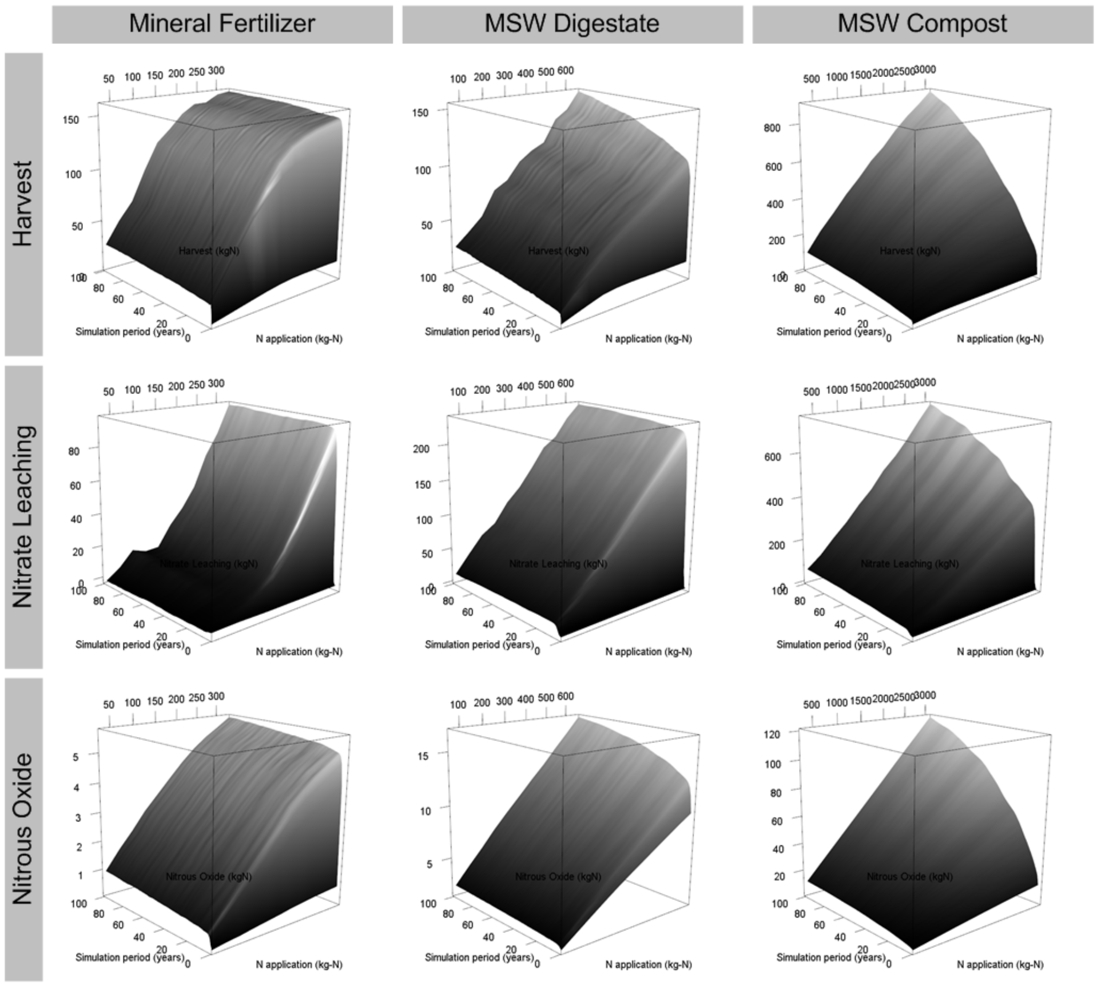

Figure 7. Response surface for harvest, nitrate leaching and nitrous oxide emissions for mineral fertiliser, anaerobically digested municipal solid waste (MSW Digestate) and composted municipal solid waste (MSW Compost) over a 100-year simulation given as the average of 64 permutations. Applications and emissions are given in per ha basis. The figure was prepared based on the result presented in Paper V.

4.3 Emission factors for organic fertilisers

Time-dependent crop yield responses and emission factors were developed by fitting a linear mixed model to the outcome for each simulation year. The R package lme4 was used to fit linear mixed-effect models (Bates and Maechler 2009). The cumulative crop yield response and emission factors were developed independently for each of the nine soil-precipitation combinations and 100 simulation years by fitting the linear mixed-effect model described in Eq 1:

$$Y_{ijk} = \alpha X + \beta + Z_{ij} + \varepsilon_{ijk} \text{ (Eq.1)}$$

where Y_{ijk} represents output variables (crop yields or emissions into the environment in kgN ha-1) for a particular combination of i (application year), j (crop) and k (fertilisation levels), X is the fixed effect variable (fertiliser application rate in kgN ha-1), α is the fixed effect coefficients, β is the fixed effect intercept, Z_{ij} is the random effect between crop and application year and ε_{ijk} is residual errors of the model. In order to capture the non-linearity of the response surface, the fertiliser application level was divided into two phases: a high response, where the amount of N applied to the field is below the rate of plant uptake, and a low response phase, where marginal crop yield gain following the application of N fertiliser ceases, as illustrated in Figure 8.

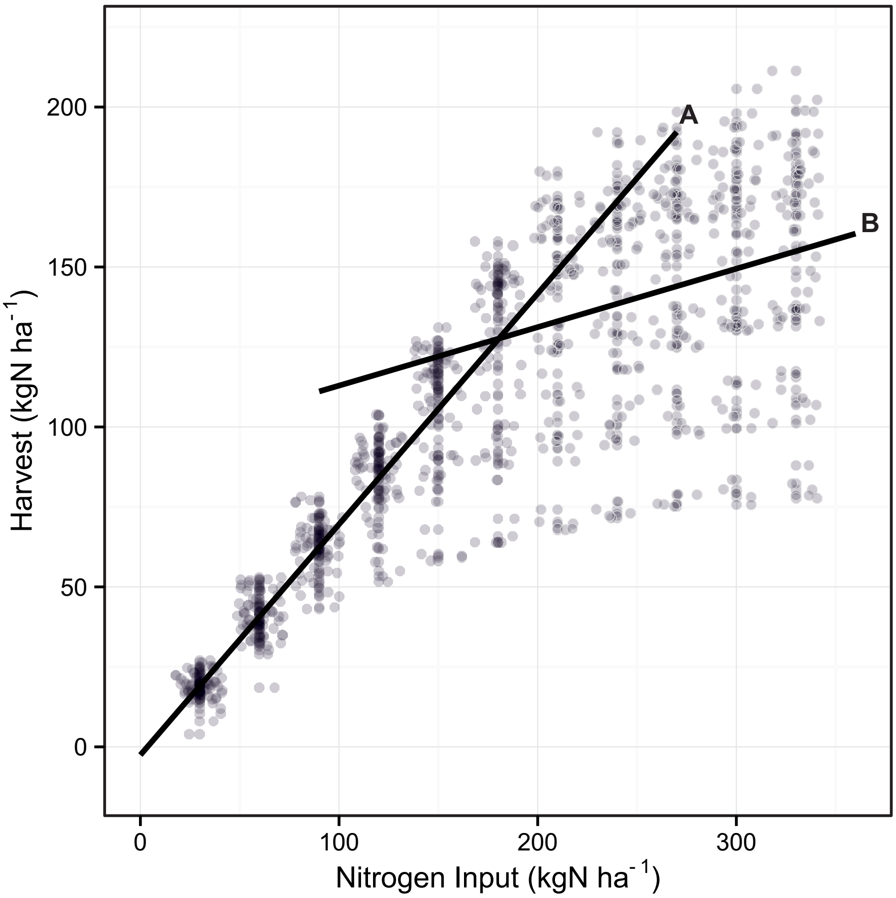

Figure 8: High and low response phases for crop yield responses to the application of mineral fertiliser. Cumulative yield response curve over a five-year simulation period for sandy loam soil under an average Danish precipitation regime was used as an example, and bold lines represent the emission factors and the dotted lines represent the associated uncertainty range. The figure was prepared based on the result presented in Paper V

Emission factors were developed for each of the nine soil and precipitation combinations for a representative mineral fertiliser (ammonium nitrate) and 11 types of organic fertiliser (Paper V, Paper VII). Variations in cumulative crop yield responses and emission factors over the 100-year simulation period are presented in Figure 9. When applied in the high response phase, none of the organic fertilisers, even the liquid digestate (MSWD and ADS in Figure 9) with high inorganic N content, appeared as being effective as mineral fertiliser. In the low response phase, the difference between mineral and organic fertiliser were less visible.

In general, organic fertiliser applications result in higher emissions of N_2O and NO_3^- into groundwater, as organically bound N continuously mineralises outside the period of plant growth and builds up inorganic N pools in the soil. The DAISY model simulates N_2O emissions and NO_3^- losses as a function of the inorganic N pool. Hence, the accumulation of inorganic N in soil resulted in elevated emissions.

As discussed further in Papers V and VII, these emissions continue to occur over a 20-year period for the majority of the organic fertiliser assessed in the present PhD project, and in the case of composted municipal organic waste, emissions continued over a 100-year period. Discrepancies among the organic fertilisers diminish over time, but differences in the rates of N_2O and NO_3^- emissions among nine soil and climate combinations persisted over time.

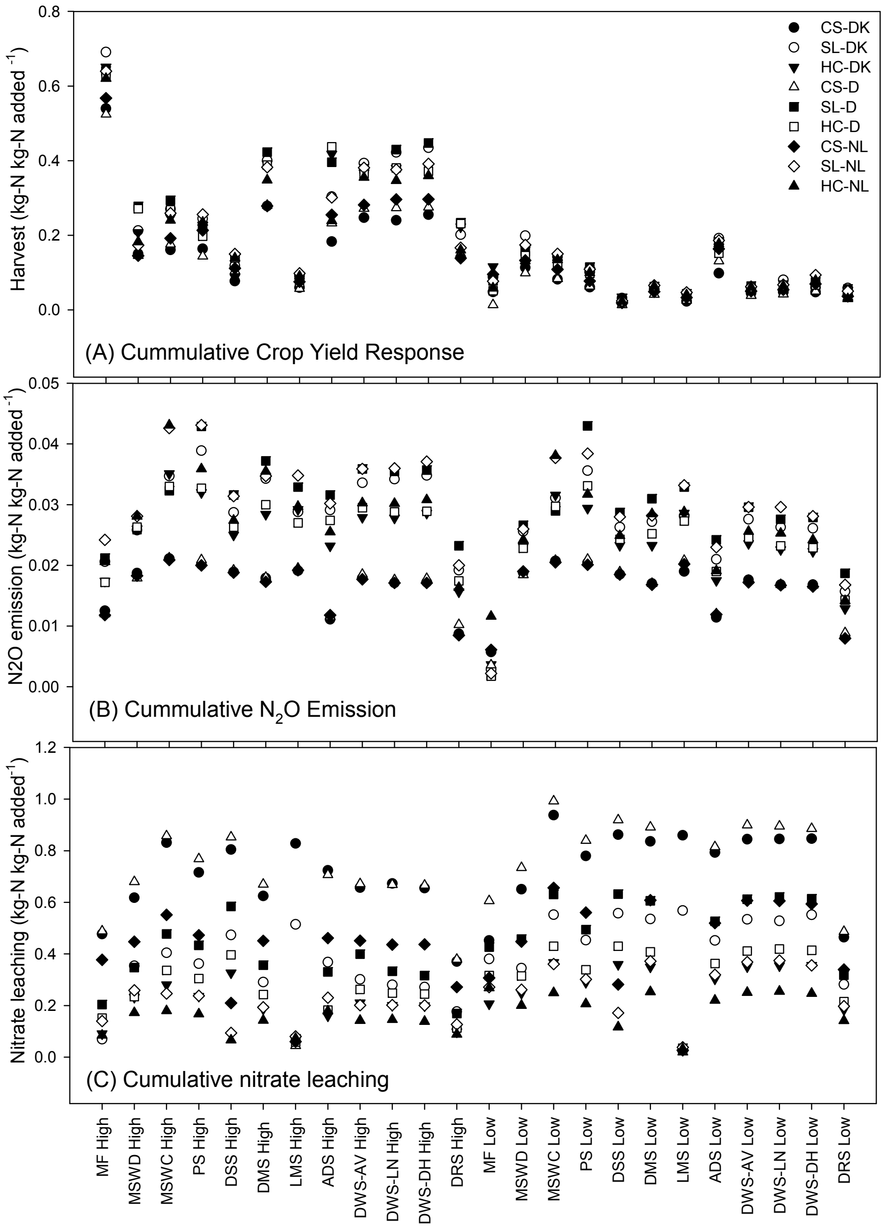

Figure 9. Cumulative crop yield responses, N_2O and nitrate emission factors over a 100-year simulation period for mineral fertiliser (MF), anaerobically digested municipal solid waste (MSWD), composted municipal solid waste (MSWC), primary sludge (PS), dewatered secondary sludge (DSS), dewatered mixed sludge (DMS), limed mixed sludge (LMS), anaerobically digested sludge (ADS), dewatered anaerobically digested sludge from three wastewater treatment plants (DWS-AV, DWS-LN, DWS-DH) and dried dewatered anaerobically digested sludge (DRS). Figure is prepared based on the result presented in Paper V and VII.

4.4 Case study: environmental assessment of the application of sewage sludge on land

In order to illustrate the shift in emission timing, and to understand the implication for environmental performance, a life cycle assessment (LCA) was applied to five treatment alternatives for sewage sludge management practice, including:

- Land application of dewatered mixed primary and secondary sludge (DMS)
- Land application of limed sludge (LIMS)
- Land application of anaerobically digested sludge (ADS)
- Land application of dewatered anaerobically digested sludge (DWS)
- Incineration of dewatered anaerobically digested sludge (INC)

Treatment and final use of 1000 kg of raw sewage sludge, generated from the municipal wastewater treatment plant, was used as the functional unit of the assessment, and the system boundary included sludge treatment, the treatment of rejected water from the mechanical dewatering process and sludge disposal or utilisation. Avoided production, applications and emissions from the field, induced by the use of conventional fertiliser on land, were also included in the assessment.

As presented in Figure 10, the land application of liquid digestate (ADS) had the highest impact potential for almost all the impact categories except for freshwater eutrophication (FE). This is due to the fact that this option would not create any loss of P via rejected water treatment and the discharge of P directly into water bodies. Anaerobic digestion coupled with the incineration of sludge had the lowest impact on global warming and three toxic impact categories, while the direct application of dewatered raw sludge turned out to be the option with the least impact on the remaining impact categories, such as terrestrial acidification, terrestrial eutrophication and marine eutrophication.

Two toxic impact categories (non-carcinogenic human toxicity and eco toxicity) exhibited an almost one order of magnitude greater impact than the rest of the impact categories, and the introduction of zinc to agricultural land dominated both impact categories. Nevertheless, the toxicity evaluation of nutritiously essential metals such as zinc remains debatable, as its toxicity depends on its concentration in environmental media rather than total mass loads.

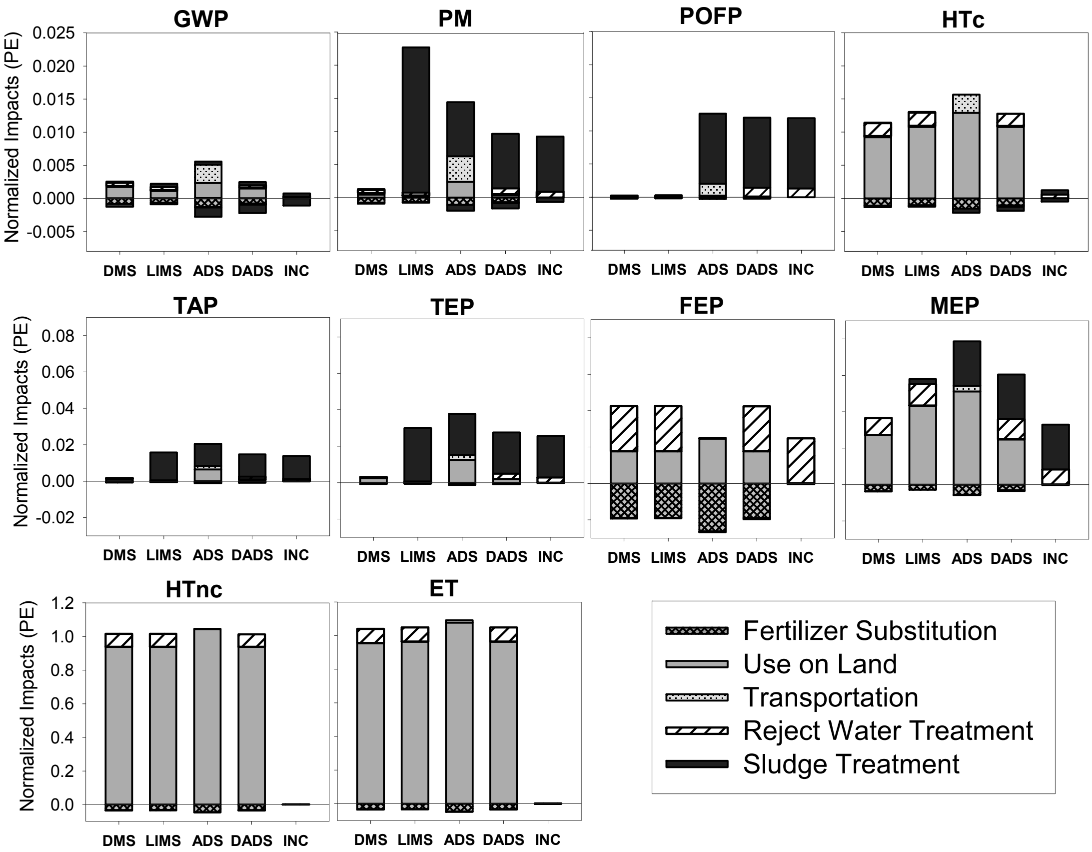

Figure 10. Characterised impacts for five treatment alternatives for global warming potential (GWP), particulate matter (PM), photochemical ozone formation potential (POFP), human toxicity (Carcinogenic, HTc), terrestrial acidification (TAP), terrestrial eutrophication (TEP), freshwater eutrophication (FEP), marine eutrophication (MEP), human toxicity (non-cancer, HTnc) and eco-toxicity (ET). Abbreviations stand for DMS: Dewatered mixed sludge, LIMS: Limed sludge, ADS: Anaerobically digested sludge, DADS: Dewatered anaerobically digested sludge, INC: Incinerated sludge (Paper VIII).

Emissions of reactive N (NH_3, N_2O, NH_4^+, NO_3^-) into the environment were the sole contributor to terrestrial and marine eutrophication potential. Also, a sizeable portion of global warming, particulate matter emissions, photochemical ozone formation and terrestrial acidification potential was associated with emissions of reactive N. For particulate matter, photochemical ozone formation, terrestrial acidification and terrestrial eutrophication, the major impacts were from reactive N emissions during the sludge treatment process, such as NH_3 evaporation from lime addition or loss of N during anaerobic digestion via denitrification or NH_3 evaporation, though these loss rates were accompanied by significant uncertainties.

Global warming and marine eutrophication potential is highly sensitive to reactive N emissions after land application, as presented in Figure 11, but when consistent local conditions such as soil and precipitation regimes are applied, the order of favourability (ADS > LIMS=DMS >DADS for GWP, ADS > DADS > LIMS= DMS for MEP) remains unchanged. However, inconsistency in reflecting local conditions in relation to the choice of emission factor could invalidate the outcome of the assessment. Hence, it is important to apply the emission factor generated under consistent local conditions, especially when the inventory is generated based on literature data.

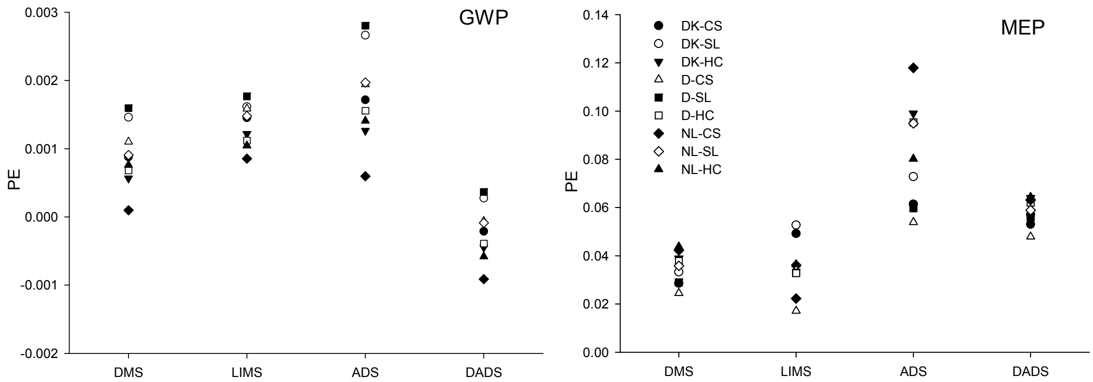

Figure 11. Variations in global warming potential (GWP) and marine eutrophication potential (MEP) caused by soil and precipitation combinations. Abbreviations stand for DMS: Dewatered mixed sludge, LIMS: Limed sludge, ADS: Anaerobically digested sludge, DADS: Dewatered anaerobically digested sludge, INC: Incinerated sludge; DK: Danish average precipitation, D: German average precipitation, NL: Dutch average precipitation, CS: coarse sand soil, SL: sandy loam soil and HC: heavy clay soil (Paper VIII).

5 Conclusions

5.1 Summary of the findings

An LCA was applied to the area of sludge management, the application of which extends from the optimisation of operational parameters to policy formulation. While providing a flexible framework to cover a range of issues imperative to sludge management practices, several research needs were identified through a review of existing LCA studies, including inconsistency in pollutant coverage, lack of site-specific data relating to biogas leakage and the omission of long-term impacts of sewage sludge land application (Paper I).

A substance flow analysis of a municipal wastewater treatment plant confirmed that besides conventionally regulated substances, such as N and P, a range of substances which pose human and eco-toxicity risks, were removed from wastewater and accumulated in sewage sludge. While representing only 1% of the total volume flow, water rejected from the sludge treatment process could represent a sizeable flow of elements through a wastewater treatment plant (Paper II). The treatment of rejected water has not been the main focus of a sewage sludge LCA as pointed out the review of existing studies (Paper I),but the omission could result in considerable underestimations of global warming, eutrophication and toxic impacts as a result of the sludge treatment process (Paper VIII).

Gaseous emissions of N_2O or CH_4 from sludge and wastewater treatment processes were evaluated via a tracer dispersion method. This allowed for the real-time measurement of unmetered gas emissions (Paper III). Higher emission rates were associated with periods of operational difficulty at the wastewater treatment plant, which could multiply the carbon footprint associated with wastewater treatment (Paper IV).

However, the collection of such site-specific data is prohibitively resource-consuming. There are already schemes to collect and even disclose to the public environmental emission data from economic entities, such as E-PRTR. The utilisation of such a scheme for the source of an LCA could provide harmonised and up-to-date emission data for pollutants. This in turn would alleviate the burden of input data collection while reducing the choice uncertainty involved in an LCA.

However, it should be noted that due to the limited pollutant coverage and omission of energy and material demand data, additional input data collection beyond existing data collection ought to be considered when conducting an LCA of wastewater and sludge treatment processes (Paper IV).

Finally, the long-term consequences of sewage sludge application on land were evaluated, using the dynamic agro-ecosystem DAISY model, which was calibrated based on C and N mineralisation rates for sewage sludge obtained from a 190-day non-leaching soil incubation test (Paper VI). Emissions over 100 years were simulated and time-dependent emission factors were developed by fitting linear regression models to the response curve for each simulation year (Papers V, VII). While the fertilising effect from the application of mineral fertiliser ceased within a short period, the effect of the organic fertiliser persisted over a prolonged time (Paper VI).

Large variations in emission factors due to local conditions were observed, especially for the case of the liquid application of sludge. Emissions of reactive N (NH_3, N_2O, NH_4^+, NO_3^-) into the environment accounted for not only all of the terrestrial and marine eutrophication potential but also a significant proportion of global warming, particulate matter emission, photochemical ozone formation and terrestrial acidification potential. The assessment of N balance beyond energy and C, often conducted for environmental impact assessment, is vital for accurately depicting the environmental performance of sewage sludge management options (Paper VIII).

5.2 Good practice for conducting an LCA

Besides filling the abovementioned knowledge gaps, several factors were identified that are relevant to good practice when conducting a sewage sludge LCA.

- **Evaluating data quality:** Emissions from wastewater treatment plants depend not only on the plant's process configurations but also on influent characteristics and day-to-day operational practices. If some emission data were taken from literature sources to fill the gap in site-specific data, or inventory data were taken from a database such as Ecoinvent, the evaluation of data quality ought to be included in the assessment.

- **Conducting a reality check:** Unlike other types of environmental modelling, the results of an LCA cannot be validated against "real" measurement data. This poses a problem in identifying errors in technical assumptions, calculations and the interpretation of results. Nonetheless, it is possible to conduct a reality check by providing mass balances of key substances such as C and N, or conducting general sensitivity analyses by calculating sensitivity ratios for all the parameters included in the modelling process.

- **Reflecting local conditions consistently:** Local conditions such as soil and precipitation regimes were found to be a factor determining the fate of C and N after the land application of both organic and inorganic fertiliser. When conducting an LCA with UOL, LCA practitioners ought to choose emission factors which consistently reflect local conditions across the treatment scenarios. This is particularly important when assessments are conducted based on literature sources.

- **Including of uncertainty assessment:** The results of an LCA are subject to uncertainties in terms of the technical modelling approach, scenario development and the assignment of parameter values (Lloyd and Ries, 2007). Though an uncertainty assessment cannot substitute for the effort exerted to collect site-specific input data, its application ought to be expanded in the area of sewage sludge management.

- **Providing adequate documentation:** The reproducibility of LCA studies depends solely on the thorough documentation of underlying assumptions and limitations. This documentation should include not only methodological (i.e. functional unit, system boundary, cut-off criteria, LCIA method) but also technical aspects of the assessment (i.e. sources of emissions and operational data, choice of parameter values and evaluation of uncertainty). However, due the word limits of academic journals, adequate descriptions were often missing from the review studied and thus posed a problem in relation to reconstructing the assessment. It is therefore recommended to provide detailed supplementary information.

5.3 Future perspectives

Despite the effort to fill the knowledge gap, this PhD work also revealed the need for further research regarding the environmental assessment of sewage sludge treatment alternatives.

- **N loss during sludge stabilisation**: Since the removal of N is not the major focus of sludge treatment processes, scientific knowledge on the fate of N during sludge stabilisation processes is limited. As N is the main contributor to almost all non-toxic impact categories, further research is needed to study the emission pathways of N loss during wastewater and sludge treatment processes and to establish correlations between N loss rates and operational conditions.
- **Dynamic modelling of a wastewater treatment plant**: When sludge is dewatered, a considerable amount of N, P and other soluble constituents seeps into rejected water and is often sent back to the head of the wastewater treatment plant. This creates a loop within the wastewater treatment plant. This process was modelled in a static manner in the present PhD study, but in order to capture fully the effect of modifying sewage sludge treatment practice to the entire wastewater treatment stream, the work should be expanded to include the dynamic modelling of a wastewater treatment plant.
- **Pathogen destruction**: Due to its origin, sewage sludge contains pathogenic organisms, and adequate inactivation of pathogenic organisms is therefore required before the utilisation of sewage sludge. While the effectiveness of sewage sludge stabilisation technologies on pathogen reduction has been studied extensively, an LCIA method to quantify potential impact due to the emission of pathogens to environment has not been established.
- **NH_3 evaporation form UOL**: The present study focused on long-term emissions, and loss as a result of NH_3 evaporation was assumed to be 15% of NH_4^+ contained in organic fertiliser. In reality, the rate of NH_3 evaporation is also affected by the chemical properties of organic fertiliser, local conditions and management practice. A further evaluation of the sensitivity of emission factors to the rate of NH_3 evaporation is needed, in order to understand fully both the short- and long-term consequence of sludge application on land.
- **Farming practice**: Besides the emission parameters examined in this study (precipitation regime, soil type, crop rotation, yearly variations in

whether data), marked differences were observed between the high and low response phases. Generally, economically optimal N fertiliser application rates, and application rates allowed by regulations, were below this critical point, and as long as farmers are making a rational choice, the N fertilisation level remains in the high response phase. However, it is often found that farmers prefer to over-apply N fertiliser due to the misconception of better crop responses to the nutrient and the aversion of risk (Sheriff 2005). Though it is highly unlikely that farmers would sabotage nutrient regulations in Denmark, if the assessment is conducted in the area the over application fertilizer is common practice, it is advisable to assess the emission factor for low response phase.

- **Effect of mixed application**: The mixed application of organic and inorganic N fertiliser could also affect the balance of inorganic and organic N in soil. Because the EU Nitrate Directive set the cap for the amount of organic fertiliser which can be applied on land, regardless of its mineral fertiliser equivalency (170 kg-N ha^{-1}), farmers are most likely to apply both mineral and organic fertiliser at the same time. In this study, the effect of combined impacts from the mixed application of mineral and organic fertiliser was not assessed. A recent review of compost utilisation suggests that the nutrient property of compost products can be better exploited when they are mixed with mineral fertilisers or consecutively applied on the same farm land (Diacono and Motemurro 2010). The evaluation of synergetic effects between organic and mineral fertiliser should be explored in future studies.
- **Improvement of soil quality**: Sewage sludge is also known to improve soil quality, such as cationic exchange capacity, soil bulk density, water holding capacity, micro-organism diversity and organic carbon content (Singh and Agrawal 2008, Martinez-Blanco et al. 2013). These benefits have not been accounted for, due to the lack of any framework for their characterisation and quantification.
- **Expansion of fate modelling**: Currently, the fate of pollutants after being deposited on agricultural land is embedded in characterisation factors used for LCAs. However, the fate of pollutants is also influenced by local factors such as soil type, climate and the actual crop being grown in the field, and the application of fate modelling should be expanded beyond C and N to other constituents of sewage sludge. An expansion of fate modelling in LCAs would enable a toxicity assessment based on exposure rate rather

than total mass load, which is currently used in LCAs. Furthermore, a toxicity assessment based on total mass load could overstate toxicity impacts, especially for metals which are essential nutrients for plants and animals and whose toxicity depends more on the concentration of environmental media.

Finally, the authors would like to re-emphasise the importance of harmonising some key technical assumptions. An LCA is an iterative process, and the coverage of pollutants and scope of studies should be expanded to reflect state-of-the-art knowledge about the environmental performance of the sewage sludge treatment process. The periodic review of scientific research and constant updates on guidelines governing pollutant coverage are anticipated, as this move would reduce unnecessary choice uncertainty and ensure the quality of LCA studies.

There are already some initiatives for establishing guidelines for LCAs of solid waste and wastewater treatment processes. The European Commission Joint Research Centre has published a technical guideline on solid waste LCAs (EC-JRC 2011), and the International Water Association has established the Working Group for Life cycle Assessment of Water and Wastewater Treatment (IWA 2013). Some of the findings from the present PhD study were incorporated in the IWA guidelines, which will be forthcoming in autumn 2014.

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I. Yoshida, H., Christensen, T.H., Scheutz, C, 2013. Life cycle Assessment of Sewage Sludge Management – a review. *Waste Management and Research* 31(11), 1083-1101.

II. Yoshida, H., Christensen, T.H., Guildal, T., Scheutz, C.. A comprehensive substance flow analysis of a municipal wastewater treatment plant. *Chemosphere*, doi:10.1016/j.chemosphere.2013.09.045

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V. Yoshida, H., Nielsen, M.P., Scheutz, C., Jensen, L.S., Bruun, S., Christensen, T.H., *submitted*. Long-term nitrogen emission factors for land application of treated organic waste as organic fertiliser. *Submitted to Environmental Modeling and Assessment.*

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VIII. Yoshida, H., Christensen T.H. Scheutz, C. Life cycle assessment of sewage sludge management alternatives including long-term impacts after land application. *Manuscript under preparation.*

In this online version of the thesis, the papers are not included but can be obtained from electronic article databases e.g. via www.orbit.dtu.dk or on request from DTU Environment, Technical University of Denmark, Miljøvej, Building 113, 2800 Kgs. Lyngby, Denmark, reception@env.dtu.dk

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